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Seasonal changes and determination of heavy metal concentrations in Veshaw river of the Indian western Himalaya

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Trace elements are a major pollutant in the river water and these pollutants are present in all components of the ecological system. Since time immemorial, the River Veshaw has been one of the important sources of water and has shaped the cultural and social values in the Kashmir Valley. This study was conducted in rural parts of South Kashmir in the Western Himalaya from February to January 2020-2022. The river Veshaw provides various ecosystem services to the local communities as well as in the upper and lower stream regions in the form of many direct and indirect ecosystem services. The river is polluted by human waste from both rural and urban communities, as well as by agricultural runoff and effluent discharges from a variety of industrial activities due to its proximity. Effluent that makes it to the river contains a variety of pollutants, some of which are trace elements that accumulate in the local ecosystem, killing off plants and animals and reducing biodiversity. Trace element levels in water and sediment were found to follow the trend as: Sangam > Khudwani > Kulgam > Nihama > Aharbal > Kingwattan. The dominance pattern of heavy metals in water was Pb > As > Cd. The overall trend showed a downward trend of heavy metals, indicating the effect of land area drainage and anthropogenic activities on the stream water. The dominance pattern of heavy metals in water was Pb > As > Cd. Heavy metals were not detected in the middle and upstream sites. The average levels of heavy metals were highest at Downstream (Sangam), with values of Cd, Pd ad As 0.0054, 0.038 and 0.038 mgL-¹. This shows that land drainage and human activities have an effect on the water in the stream.

KEYWORDS

seasonal changes, heavy metal, cadmium, lead, arsenic, himalaya

1 Introduction

The pollution of the aquatic ecological system with trace elements has become a worldwide ecological problem in recent years. These trace elements are indestructible and most of them have toxic effects on aquatic organisms (MacFarlane and Burchett, 2000; Storto et al., 2021). Among various environmental pollutants, heavy metals are of particular concern due to their potential toxic effects and ability to bioaccumulate in aquatic ecosystems (Fernandes et al., 2006; Khan N. A. et al., 2020; Bhat et al., 2022). Trace elements in general, and heavy metals in particular, are significant toxicants found in surface water (Buragohain et al., 2010). Trace elements are one of the most serious pollutants in the natural environment due to their toxicity, persistence, and bioaccumulation problems (Pekey, 2006; Nouri et al., 2006). Many scholars have studied the trace element contamination of surface water in different locations in India (Ram and Singh, 2007; Tiwari et al., 2016; Lohani et al., 2008). Trace element pollution has been assessed by the distribution of particle size and the organic content present in the water system (Saif and Khan, 2017; Bano et al., 2018; Prieto et al., 2018). The bioavailability of trace elements depends on the concentration of anions, chelating agents in the water, pH, and the presence of absorptive sediments. Ali and Khan, 2018 suggested in their study that the increase in trace elements in the surface water was due to the addition of effluents released from industrial and commercial areas.

Heavy metal (HM) contamination has piqued the world's attention due to its toxicity to living organisms through the bioaccumulation process (Ekmekyapar et al., 2012). HMs are typically classified as metals or metalloids with a higher elemental density. These are subdivided on the basis of density, weight or atomic number (Teixeira de Souza et al., 2021; Bhat et al., 2022; Kumar et al., 2022). The new standard, stating that HM should be mineral mining (Ali et al., 2019; He et al., 2021) According to some researchers the main sources of heavy metal concentration (HMC) in China are agriculture and manufacturing, as a large amount of waste is produced in these activities, posing significant health risks to society on a large scale (Sun et al., 2010; Ohiagu et al., 2020; Karmakar et al., 2021; Bhat et al., 2022). In addition to the above, cosmetics and chemical fertilisers contribute to heavy metal pollution (Kanwar et al., 2020). Intriguingly, heavy metals released by vehicles have been deposited on plant leaves and the surface of soil (Harrison et al., 1981; Vardhan et al., 2019; Malunguja et al., 2022). Studies have demonstrated the occurrence of different hazardous HMs in the soil or farm areas (Turer and Maynard, 2003; Bhat et al., 2022). Recent studies conducted on untreated effluent water have been proven to be a substantial source of HM polluted water and other areas of the world (Tauqeer et al., 2022). Sewage water has been linked to the presence of potentially harmful elements like Cd, Cu, Ni, Cr, Pb and Zn in soil, plants, and foods (Natasha et al., 2022).

The unplanned and unregulated development of urbanisation during the last two decades has resulted in the degradation of environmental health, particularly in developing countries like India (Rauf et al., 2009; Madhav et al., 2020; Bano et al., 2022). A number of anthropogenic activities may alter the dynamic properties of lotic ecosystems by adding heavy metals to the water bodies (Cortecci et al., 2009; Bhat et al., 2011; Hasan et al., 2021; Khan et al., 2022). The presence of heavy metals in water is considered devastating and disturbs the aquatic system due to their toxicity, persistence, non-degradability, and bio-accumulation properties. All these factors are leading to serious anomalies in the aquatic ecosystem (Yang et al., 2009; Khan A. H. et al., 2020; Rather RA. et al., 2022). Trace metals may be naturally present in the biosphere and their fast release into the environment is augmented by human activities. The chemical cycling of trace metals is intricate due to the variety of factors (biotic and abiotic) involved in the manifestation of metal behavior, such as bio-chemical processes, hydrology, climatic factors, land use, and metal characteristics (Husain Khan et al., 2020; Mazhar et al., 2020; Khan et al., 2021). (Damian et al., 2019; Padder et al., 2021; Rather et al., 2022c; Padder et al., 2022) looked at the ecological value of trace metals in fresh water systems and the level of pollution in the water of the Kashmir Himalayas.

The present study is based on the analysis of seasonal changes in monitoring the quality of drinking water in the Veshaw River in the Indian Western Himalayas. The purpose of conducting this study was to improve the quality of drinking water in that area on the basis of the recommendation of the study. The Veshaw river is located adjacent to the agriculture and horticulture area and small workshop and mini-industrial units. The majority of these wastes and effluents are discharged into the environment as a result of these activities. These various forms of affluent discharged into the river are major concerns for changing the ecological system of the river. The Veshaw river is a major source of water for domestic, agricultural, and industrial uses in the south of Kashmir. Water from the Veshaw river is being used for various purposes that may expose the users to some health hazards, hence the need to determine the level of heavy metals it contains for the safety of the users. The main objective of this study is to investigate the presence and to examine the average concentration of heavy metals in the water of the Veshaw River; this involves investigating the possible natural sources and impacts of anthropogenic activities on the water quality.





FIGURE 2

Sampling locations during collection of water samples from Veshaw river [(A):Kongwattan; (B)Aharbal; (C):Nihama; (D):Kulgam; (E):Khudwani; (F):Sangam].

2 Materials and methods

2.1 Sampling procedure and analysis of heavy metals

Three different locations were used to collect water samples for testing the quality of the rice water, viz., Site-I and II (Kongwaton $33^{\circ}62'80.3''N,\ 74^{\circ}74'08''E$ and Aharbal $33^{\circ}38'45.4560''N$ 74°47'50.4696"E, an upstream site); Site-III and IV (Nihama $33^\circ 38'8'' N \ 74^\circ 53'34'' E$ and Kulgam $33^\circ 38'24'' \ N \ 75^\circ 01'12''$ E, a midstream site); and Site V and VI (Khudwani 33.7074° N, 75.1054° E and Sangam 33.828,217°N 75.070842°E, a completely downstream site) mentioned in Figures 1, 2. The locations were chosen depending on the impacts of anthropogenic activities or on the basis of the observation of the visual analysis of pollution sources like agriculture activities, horticulture activities, human habitation, commercial and other anthropogenic and livestock pressures along the banks of the Veshaw river. From these survey sites, the collected samples were analysed to identify seasonal fluctuations (Rather et al., 2022b; Rather et al., 2022e). The samples were collected at an interval of 2 months in the years 2020-2022. Five water samples were collected from each sampling site, and a total of 30 water samples were analysed in the laboratory. The seasonal sampling was carried out, and the samples were stored in sterilised bottles. In order to analyse the chemical features of the water, it was immediately refrigerated at -4°C and transferred to the lab. In addition to sorting data by season, the results were analysed to identify seasonal fluctuations.

2.2 Trace element estimation in water

A total of 500 ml of water was collected up to a depth of 30 cm and immediately transported to the laboratory where the samples were filtered and acidified with a few drops of concentrated HNO3 before being preserved in polythene bottles for subsequent trace element analysis using an Inductively Coupled Plasma Optical Emission Spectrophotometer (Varian Vista MPX, 720). Among the estimated trace elements are the trace elements (APHA. 2005; Rather et al., 2022a).

Principle: Equipment for ICP optical emission spectrometry consists of a light source unit, a spectrophotometer, a detector, and a data processing unit. An aqueous sample is converted to aerosols via a nebulizer. The aerosols are transported to the inductively coupled plasma, which is a high temperature zone (8,000-10,000°C). The analytes are excited to different (atomic and/or ionic) states and produce characteristic optical emissions. These emissions are separated based on their respective wavelengths and their intensities are measured. The intensities are proportional to the concentrations of analytes in the aqueous sample. Total elemental concentration is analysed by ICP-OES. The Merck-Certipur reference trace element standards were used to calibrate the ICP-OES and the detection limit for each trace element was also calculated before analysing the samples.

The achieved detection limits are: arsenic (As) 0.80; lead (Pb) 0.29; cadmium (Cd) 0.11 mg kg^{-1} . The developed method is

applied for the determination of hazardous metals in the water of the river Veshaw.

2.3 Statistical procedures

The mean values of parameters were presented and analysed using descriptive analysis. Using the four distinct seasons of the year, we were able to categories the information into 4 seasons: spring, summer, autumn, and winter. Correlation matrix through XL-stat after two-way ANOVA SPSS statistic version 23 was used to collect the data. ANOVA was used to examine the differences between all of the data points (ANOVA).

3 Result and discussion

Many harmful substances, including heavy metals and other trace elements, can be found in surface water. Many regions of India have had their surface water reported to be contaminated with trace elements (Lohani et al., 2008). Particle size distribution and the organic content in the water system are considered to be contributors to trace element pollution (Simokawa et al., 1984). All trace elements in the present investigation revealed substantial seasonal and spatial variability.

3.1 Cadmium

In river water, cadmium (Cd) is mostly present as CdCO₃ species. In recent decades, there has been an increase in the production and use of Cd and its compounds. Cd enters river water from industrial discharges, mining wastes, and atmospheric deposition due to the combustion of fossil fuels (Syres et al., 1986; Patel et al., 2018). Another source is washout from agricultural land because some fertilisers contain Cd levels of even up to 40 mg/kg (Kumar et al., 2022). One of the major sources of diffuse Cd pollution is the production of inorganic fertilisers from phosphate ores (Paul et al., 1994; Rashmi et al., 2020). The major environmental Cd sources are Zn and Pb ores as it occurs naturally in their sulphide ores. Commercially, it is produced as a by-product of the Pb and Zn smelting processes (Stewart et al., 2022) Raw Zn, Zn alloys, and Zn compounds may have high concentrations of Cd, so that it may enter the river water from solid wastes as well (Emoyan et al., 2005). At sewage treatment plants, the presence of Cd in waste is a matter of great concern because sewage sludge contaminated with Cd becomes unfit for use in fertilising soils (Lokhande and Sathe, 2001; Penido et al., 2019). The sites of greatest Cd accumulation in the body are vital organs like the liver and kidney. After gastrointestinal absorption, Cd is concentrated in the kidney. Cd poisoning causes nephrotoxicity (kidney damage). Other toxic effects of Cd exposure include a decrease in haemoglobin concentration, erythrocyte destruction, an increase in blood pressure, liver damage, and male sterility (Aggarwal et al., 2000; Priyadarshanee et al., 2022).

The concentration of cadmium in the river Veshaw is shown in Figure 3. The cadmium concentration was found below the detection limit in the upstream and in the middle stream during the entire study period of 2020-2022. The lowest amount of Cd was found in the middle stream (Kulgam) at 0.0013 mgL-1. In the summer, the maximum amount of Cd was found in the downstream (Sangam) area at 0.0091 mgL⁻¹. In the Veshaw river, the cadmium concentration was found below the detection limit upstream at an undisturbed site (Kongwattan and Aharbal) and in the middle stream at Nihama during all the seasons (Figure 3). Cadmium was detected in the middle and downstream during all seasons. The overall trend showed a significant downstream increase (p 0.05) in the cadmium level of water. The increase in the cadmium concentration downstream may be attributed to increased anthropogenic sources of cadmium from the riparian zone in the form of increased domestic sewage, dumpsites, runoff from workshops, agricultural fields, and sanitary pipelines The results of the present study are in agreement with the results of Aggarwal et al. (2000), Lokhande and Sathe (2001), and Amuah et al. (2022). Seasonal variation of the collected samples revealed the highest concentration of this metal in the summer and autumn seasons, coinciding well with the low discharge during these seasons. A similar seasonal trend was observed by Mondol et al. (2011) around Tejgoan, Bangladesh. The study conducted by Yao et al. (2014) also reported the seasonal difference in the cadmium concentration in the rivers of China, with a high concentration during the dry (summer) season and related it to the background and long-term accumulation in the environment. Other related seasonal trends for Cd have been reported by Emoyan et al. (2005) in the river Ijana, Nigeria; Kar et al. (2008) in the river Ganga, West Bengal, India; and Buragohain et al. (2009) in the Dhemaji stream, Assam, India. In addition, Paul et al. (1994) also reported higher levels of Cd in the summer, while Kaushik et al. (2009) observed the lowest and highest Cd concentrations during the summer and rainy seasons in the river Periyar, Kerala and the river Yamuna, Haryana, Gwalior, respectively. The concentration of Cd at this point was found to be below the detection limit (BDL) in all the seasons. Sangam downstream showed high levels of Cd during the entire study period as this site receives heavy loads of sewage and effluents from a variety of small-scale industrial units, which dump their wastes directly into the river without any treatment. Cooper et al. (1978) and Aggarwal et al. (2000) have reported an increase in Cd concentration in water with an increase in the quantity of sewage, while high Cd content in



water receiving commercial, domestic, and industrial effluents has been reported by Lokhande and Sathe (2001). Cd also finds its way into water from agricultural runoff as fertilisers are known to contain Cd impurities (Syres et al., 1986).

3.2 Lead (Pb)

Inorganic Pb occurs in water as carbonates and hydroxides. Major anthropogenic inputs of Pb in water include industries, mining, smelting plants, and vehicular inputs. It also enters into water bodies from sewage sludge. Pb is used in the production of lead acid batteries, solder, alloys, cable sheathing, pigments, rust inhibitors, ammunition, glazes, and plastic stabilizers. Tetraethyl and tetramethyl Pb are important because of their extensive use as antiknock compounds in petrol in many developing countries. Mild Pb poisoning can cause anaemia, headaches, sore muscles, fatigue, and irritability. Acute Pb poisoning causes severe kidney, liver, reproductive, and central nervous system dysfunction (Strehlow and Barltrop, 1987; Balali-Mood et al., 2021). It damages RBCs and delta-aminolevulinic acid dehydratase with heme activity, interferes synthesis, inhibits haematopoiesis and produces adverse effects on blood vessels (Shafiq-ur-Rehman, 2010; Rahimpoor et al., 2020). Pb can also cause osteoporosis by replacing calcium in bones (Ferner, 2001). The role of Pb in causing mental retardation, particularly in children, is also well documented by many researchers (Ferner, 2001). Pb is present in surface water as a result of its dissolution from natural sources, but primarily from household plumbing systems in which the pipes, solder, fittings, or service connections to homes contain Pb. Polyvinyl chloride (PVC) pipes also contain Pb compounds that can be leached from them and result in high Pb concentrations in surface water. The amount of lead dissolved from the plumbing system depends on several factors, including the presence of chloride and dissolved oxygen, pH, temperature, water softness, and standing time of the water Tam and Elefsiniotis, 2009). The concentration of the lead content in the Veshaw stream is shown in Figure 4. One striking difference found in the lead concentration was that the cadmium concentration was found below the detection limit at upstream (Kongwattan and Aharbal) during the entire study period in all the seasons. In the winter, the middle stream (Nihama) had the lowest average value (0.004 mgL⁻¹), and in the summer, the downstream (Sangam) had the highest average value (0.046 mgL⁻¹). In the present study, lead was found below the detection limit upstream at Kongwattan and Aharbal during all the seasons (Figure 4). Afterward, its concentration revealed a downward trend, with the maximum mean value observed in



the lower reaches of a stream (Sangam). The highest concentration was observed in the summer season, followed by the autumn season, and the lowest was observed during the winter season. The highest concentration of lead in summer and autumn could be due to less dilution owing to the low discharge of the stream. A similar trend within the river was observed by Quan et al., 2022). Increased lead content downstream may be due to the inflow of waste from garbage dumps, domestic sewage, transportation, vehicle workshops, and industrial discharge from industrial units within the catchment zone of the stream. Our findings are in agreement with the previous studies carried out by Kaur. (2012); Baralkiewicz et al. (2014); and Appiah-Adjei et al., 2019). Koul et al. (1988) also observed the lowest Pb content in spring and the highest in summer and autumn in the Himalayan lakes of Kashmir. According to Kaur (2012), Pb was added to the river Veshaw from wastes, agricultural activities, transportation activities, and motor workshop effluents.

3.3 Arsenic

Arsenic (As) is the 20th most common element in the earth's crust and is associated with igneous and sedimentary rocks. Although elemental arsenic is not soluble in water, its salts exhibit a wide range of solubilities depending on pH and the

ionic environment. As can exist in both inorganic and organic forms. Arsenic (As+3) and arsenate (As+5) are inorganic forms of arsenic, whereas monomethylarsenic acid, dimethylarsenic acid, arsenobetaine, arsenocholine, arsenolipids, and arsenosugars are organic (Elci et al., 2008; Kalia & Khambholja 2015). Arsenicals are used commercially as alloying agents in the manufacture of transistors, lasers, and semiconductors, as well as in the processing of glass, pigments, textiles, paper, metal adhesives, wood preservatives, and ammunition. As concentrations in natural waters range between 1 and 2 g/L (Hindmarsh and McCurdy, 1986; Rohanifar et al., 2018). As concentrations rise in areas with volcanic rock and sulphide mineral deposits, as well as in areas with human activity (Hood and Bishop, 1972; Gibson and Gage, 1982; Hindmarsh and McCurdy, 1986; Wang et al., 2019).

Arsenic was found below the detection limit in the upper reaches of the upstream (Kongwattan and Aharbal) in all seasons. Figure 5. During the summer season, the middle stream (Nihama) had the lowest As concentration (0.005 mgL^{-1}) and the highest (0.042 mgL^{-1}) downstream (Sangam), followed by Khudwani (0.038 mgL^{-1}). In the downstream, the middle stream (Nihama) had the lowest As level (0.009 mgL^{-1}) and the downstream (Sangam) had the highest As level (0.038 mgL^{-1}). The overall increasing downward trend was statistically significant during all the seasons. summer recorded the highest arsenic value, followed



by autumn and spring, respectively. As levels in natural waters are typically between 1 and 2 mgL⁻¹ (Hindmarsh and McCurdy, 1986), Human habitation and the presence of sulphide mineral deposits (found in volcanic rock) lead to increased concentrations of As (Hood and Bishop, 1972; Gibson and Gage, 1982; Hindmarsh and McCurdy, 1986). As has been detected at all locations except those further upstream. The amount of As found upstream (in Kongwattan and Aharbal) was higher than in other areas, while being much below the detectable limit. Data suggests that the Veshaw's middle stream and downstream get a disproportionate share of the river's toxic waste when compared to the river's other catchment areas. As such, concentration has also been shown to be higher in the summer and fall than in the winter and spring. It suggests that middle and down stream area of the Jhelum river are receiving more toxic wastes as compared to the catchment area of the Veshaw river. Moreover, As concentration has been recorded more in summer and autumn compared to winter and spring seasons.

3.4 Correlation between the heavy metal parameters

To observe the relationship between heavy metal parameters during the different seasons, a correlation matrix based on the Pearson correlation coefficient (2-tailed) was employed between important parameters of water samples Figures 5A,B.

3.4.1 Pearson correlation between the heavy metal parameters

Figure 6 depicts the results of an investigation into the relationship between heavy metal water quality and cadmium, lead, and arsenic, which have all been found to have a significant positive correlation with one another. This is the degree of association among the following water quality factors. Figure 6A also shows that arsenic has the highest correlation (0.989**) with cadmium, while lead has the lowest correlation (0.872*) with cadmium. The correlation among pH, EC,WT, Turbidity, N, P, K, TH, Ca, Mg, Na, Fe, Cu, Zn, Pb, Cd and As





contents in water Coliform were found statistically highly significant at ($p \le 0.05$) confidence.

The regression statistics of the physicochemical data set of the Veshaw stream are summarized in Figure 7. The connection with heavy metal water quality variables was significantly equal to each other. Cadmium exhibited a similar proportionate association with Pb and As. The relative influence of the components was evaluated as follows: Cd > Pb > As. The regression findings using all station

data indicated the significant water quality characteristics at each location. At each site, the water quality parameters are affected by the level of heavy metals in the Veshaw river water. Related works were performed by authors (Hong et al., 2015; Jung et al., 2016; Seo et al., 2019) who similarly spatially categorised the Nakdong River as the up-and-midstream and mid-and downstream portions at a non-weir station between Dasa (Gangjeong-Goryeong weir) and Nongong (Dalseong weir) locations using cluster analysis.

4 Conclusion

The Veshaw River shows an increasing trend of pollution from its upper to lower reaches. The upstream water quality analysis reveals an oligsaprobic (low pollution load) nature, whereas the -mesosaprobicc nature (critical pollution load) of water in the middle and downstream reaches The results of the present study have indicated that trace elements have made their entry into the aquatic biological ecosystem and are a matter of great concern. Trace element levels in water were found to follow the trend: Sangam > Khudwani > Kulgam > Nihama > Aharbal > Kingwattan. Comparing the water quality parameters to the Indian drinking water quality standards shows that the concentrations in Khudwani and Sangam, which are near the end of the river Veshaw, are too high.

- Concerned authorities at national, state and district levels should take immediate measures (such as a complete ban on the discharge of untreated industrial effluents and municipal sewage into the river) for the restoration of the river Veshaw.
- In the area where the Veshaw river starts, there should be rules about how effluents from human activities can be made and where they can be dumped.
- Stream management activities should be prioritised in urban, rural, industrial, and agricultural areas.

Data availability statement

The original contributions presented in the study are included in the article/supplementary material, further inquiries can be directed to the corresponding authors.

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Author contributions

RR, SA provided the conceptualization and Methodology; Visualization and Investigation by SA, SS; Software SP, TB; Validation and framed the manuscript by RR, SP, SM, FL, ZB performed the analysis and formatting; RR, SA, TB, and SP helped data collection data analysis; SS helped in the initial draft of the manuscript text and revised and commented on the manuscript critically.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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